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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Rapid Evolution of Aerosol Particles and their Optical Properties Downwind of Wildfires in the Western U.S." *by* Lawrence I. Kleinman et al.

Anonymous Referee #3

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Kleinman et al. presents measurements of aerosol collected during the BBOP field campaign in 2013. The analysis focuses on the evolution of aerosol downwind of the emission. The measurements range from a half hour to several hours old. At this timescale the authors identify the main trend as an increase in MSE along with other trends in aerosol composition and ion balance. These measurements represent an important airborne dataset sampling fresh wildfire smoke in the western US. The topic is appropriate for ACP and publication of these data and analysis is important in their own right and for comparison with other recent field campaigns (WECAN and FIREX-AQ) which also focused on sampling fresh wildfire smoke in western US.

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Discussion paper

The presentation of results this manuscript is complete but the discussion of the data is qualitative and is organized around short descriptions of the figures. Some panels in the figures are not mentioned in the text. Much of the results and discussion feel like a draft manuscript. I suggest that the author revise these sections.

Specific comments: Regarding the importance of coagulation in the aging of these plume, the manuscript oscillates between stating the coagulation is the main explanation for the increase in particle size and MSE downwind of the fires, and remaining agnostic about its contribution compared to evaporation/condensation. However, a coagulation calculation is presented for only one of the Lagrangian experiments (for the highest smoke concentration observed) and it suggested that coagulation can only account for a fraction of the increase in MSE. It seems that by performing the coagulation calculation for all of the plumes sampled here, the authors may be able to place a meaningful limit on the contribution of coagulation to the increase in MSE.

This same team used the BBOP dataset to analyze the formation of tarballs in these same plumes (Sedlacek et al. 2018). I expected to see some connection to tarballs in this analysis. Do the tarball sizes increase along with the rest of the aerosol? Are changes in the OA O:C ratio correlated with the tarball fraction? Some more connection would be informative as tarballs should account for significant fraction of the OA.

Technical Comments: Page 2 line 9 -11: Abstract states the main process is evaporation and condensation. Coagulation is not mentioned. implies disagreement with conclusions which are ambivalent about coagulation vs evaporation/condensation Page 2 line 11: typo ‘transects’ Page 7 ine: 203 ‘thermal denuder’ -> ‘thermodeunder’ Page 12 line 375: missing a closing parenthesis Page 14 line 417: the sentence could much more precise Page 14 line 436. Missing period in et al. Page 14 lines 441-443: This sentence is confusing. Maybe the higher values come from hot spots (flaming spots?) in the burning area. Or are these separate smaller fire? Figure 3: scattering is misspelled. Show the location of the fire and an arrow showing the general wind direct would help orient the read more quickly. Longitude is capitalized in the caption Page

15 line 462 – 470: Please consider rewriting this paragraph. The reasoning is implied rather than explicit. The data discussed in the text (NOx, NOy , CPC) is not shown on the figure (scattering). CPC25 is not defined, although can be assumed to be from the TSI CPC3025 instrument. And that coagulation leads to a loss of small particles between the two transects. Page 15 line 465: CPC25 is not defined. Page 15 line 474: "and more extended in altitude." This seems likely but not discerned from the data presented. Figure 4: Consider coloring the bars for the two Lagrangian experiment different colors. Page 15 lines 475-476: To my eye, the divergence in between CO and scattering is more apparent in the North plume and the south plume. Maybe this should be discussed in the text/ or the north and south plumes could analyzed separately. Page 16 line 482: missing period Page 16 line 493: "Other Properties vary between North and South." Please expand a bit. What properties and how do they vary? Figure 5: Please add some more explanation of the figure to the caption. Page 16 lines 479- 484: For the second Lagrangian experiment, the transects are out of order. Please mention this in the text. Page 16 lines 504 -510: What was the purpose of transects 14 and 15? They are shown in figure 7, but not mentioned in the text. Transect 13 shows a kink in figure 20. I'm curious if transect 13 was not completely downwind of all of the emission for this fire and perhaps transect 12, 14, or 15 would be better for initializing the coagulation model. Figure 7: extra period in the caption. Please change the closing bracket to the parenthesis. Page 17 line 541 and 543: Age is unnecessarily capitalized. Repeated through the remained of the manuscript. Figure 11: Only panels c and d are briefly discussed in the text. Outliers such are simply explained away "as within the scatter". Panel b shows effective MAC, should one interpret the 821b flight to be heavy influenced by BrC as the MAC is much higher than expected from BC + lensing alone? This figure needs more detailed discussion in the text. Page 19 lines 590 -592: Is this also the case for flight 730a? It seems to be similarly outside the rest of the data. Page 21 line 667: typo, 'burn' should be 'burning' Figure 20: Is a linear fit to these data meaningful? Why are the intercepts held to zero? I would consider the transect 12 to be the initial transect, and constrain the linear fit to

go through transect 12. This interpretation of the figure suggests that coagulation can only account for a small fraction of the growth in MSE. The FINS MSE trends below the 1:1 line with increasing age for transects 10-7. Does this suggests that the FINS size distribution (even when extrapolated) is missing a significant fraction of the scattering or is this due to a changing index of refraction of OA as the sampling move downwind? Page 25 line 15: The coagulation calculation conclusion (factor of 2 increase MSE) rests on one data point from transect 13. If one initialed the calculation from transect 12, one might conclude a much smaller fraction of the MSE increase could be due to coagulation.

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